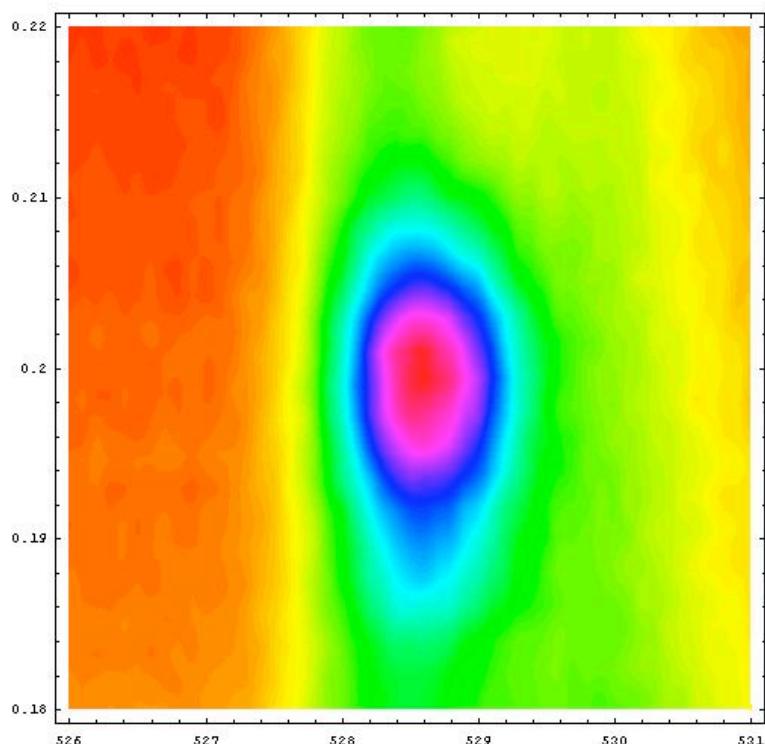


Hole crystallization in the spin ladder of $Sr_{14}Cu_{24}O_{41}$

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Luc Venema, *U. Groningen*
Hiroshi Eisaki, *AIST, Tsukuba, Japan*
Eric Isaacs, *Argonne National Laboratory*
George Sawatzky, *University of British Columbia*

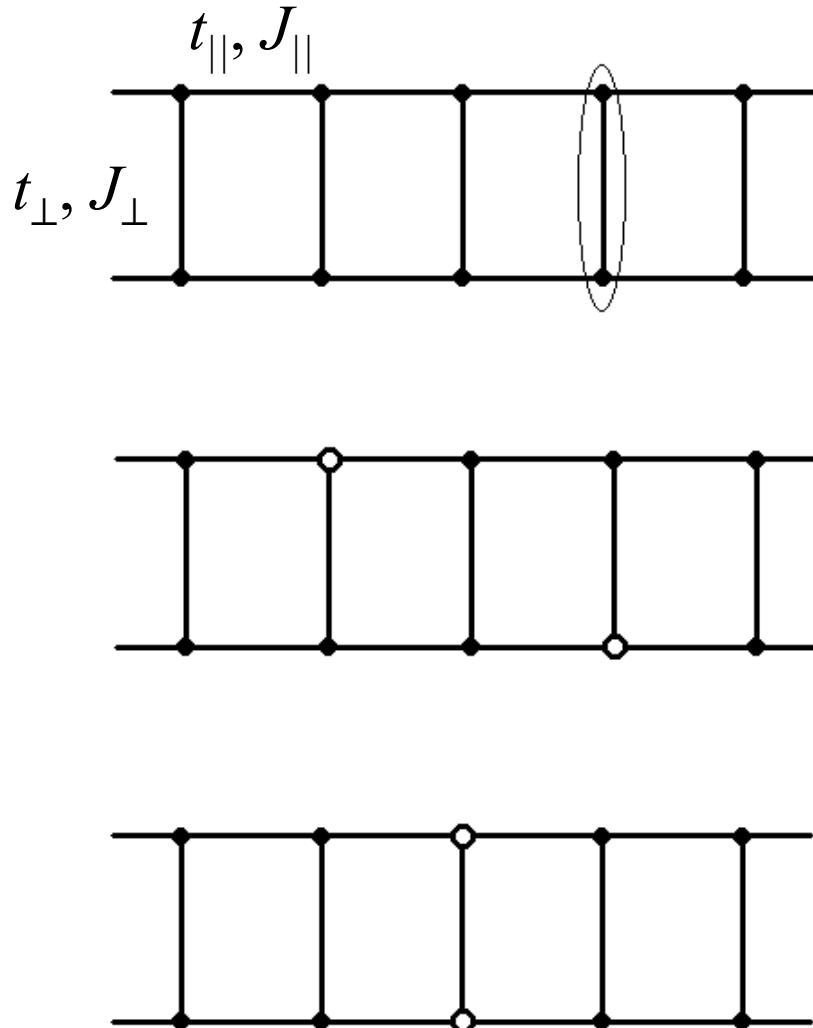
Acknowledgements to:

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Spin ladders

E. Dagotto, J. Riera, and D. Scalapino, *Phys. Rev. B*, **45**, 5744 (1992)
E. Dagotto and T. M. Rice, *Science*, **271**, 618 (1996)

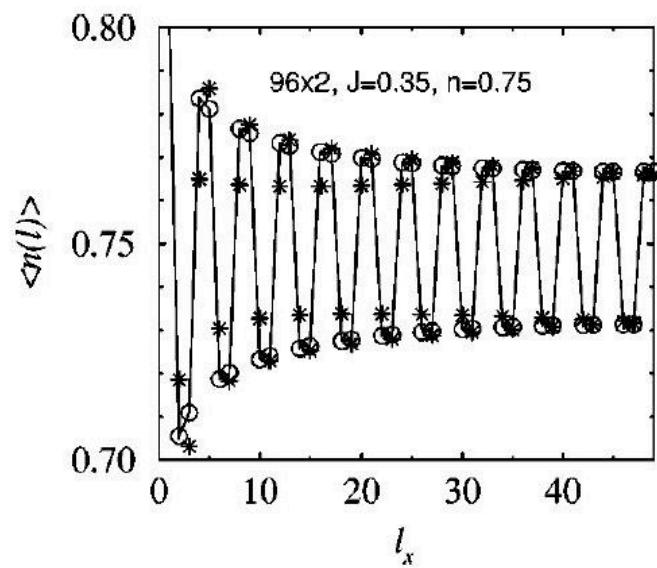
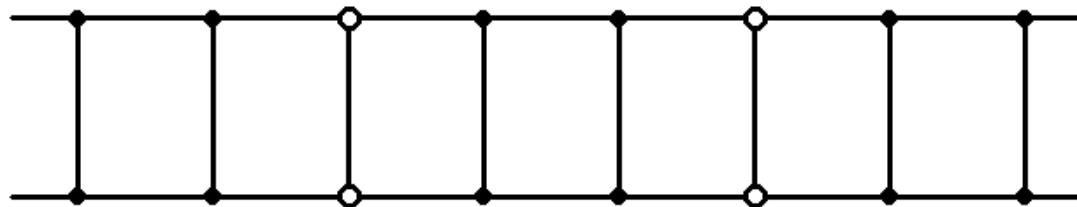


- Spin liquid (exponential decay in correlation)
 $J_{\perp} \gg J_{||}$
- Singlets across the rungs
- Doped hole breaks a singlet (costs $\sim J_{\perp}$)
- Holes bind into pairs
- Superconductivity without phonons,
 $D \sim d_{x^2-y^2}$ [M. Sigrist, *PRB*, **49**, 12058 (1994)]

Spin ladders

E. Dagotto, J. Riera, and D. Scalapino, *Phys. Rev. B*, **45**, 5744 (1992)
S. White, I. Affleck, and D. Scalapino, *Phys. Rev. B*, **65**, 165122 (2002)
S. Carr, A. Tsvelik, *Phys. Rev. B*, **65**, 195121 (2002)

Superconductivity not automatic – models also reveal a “charge density wave”

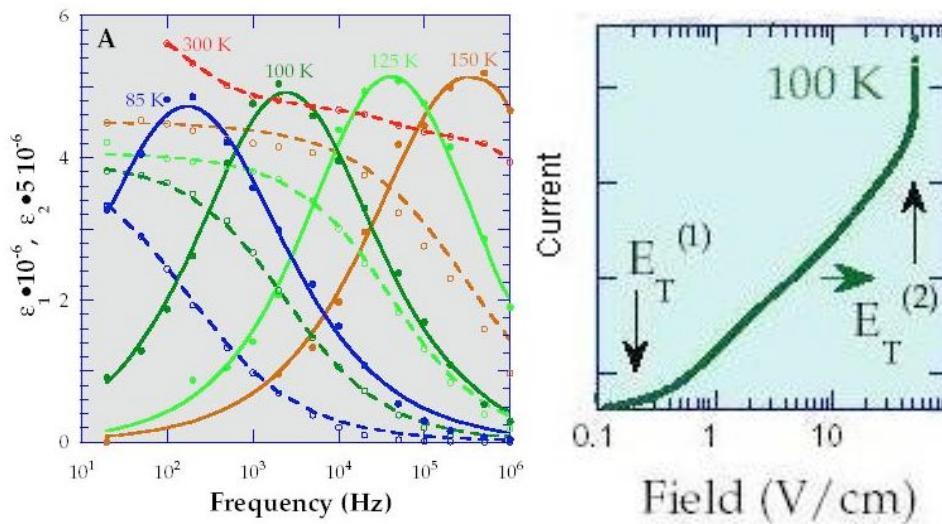


- holes “crystallize”, drive system insulating
- Particularly stable for d rational (commensurate)
- Almost a CDW: power law correlations $1/|n-n'|^{K+r}$. Friedel oscillations easily induced.
- Reminiscent of ordered stripes vs. SC in perovskite cuprates
- *Not high T_c , but worthy of study in its own right*

$Sr_{14-x}Ca_xCu_{24}O_{41}$

$$x = 0 \Rightarrow \text{Sr}^{2+}, \text{O}^{2-} \Rightarrow \text{Cu}^{2.25+}$$

- isoelectronic to perovskite cuprates
- 6 holes / formula unit
- ladder has larger electronegativity:
- 5.2 holes on chain, 0.8 holes on ladder ¹
- $d_{\text{chain}} = 0.52$, $d_{\text{ladder}} = 0.057$

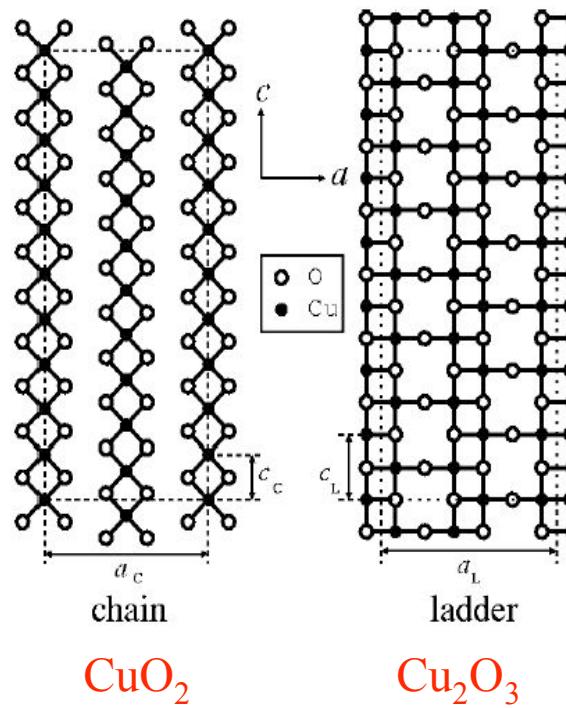


¹Osafune, *PRL*, **78**, 1980 (1997); Nücker, *PRB*, **62**, 14384 (2000)

²Uehara, *J. Phys. Soc. Jap.*, **65**, 2764 (1996)

³Blumberg, *Science*, **297**, 584 (2002); Gorshunov, *PRB*, **66**, 60508 (2002)

⁴Vuletić, *PRL*, **90**, 257002 (2003)

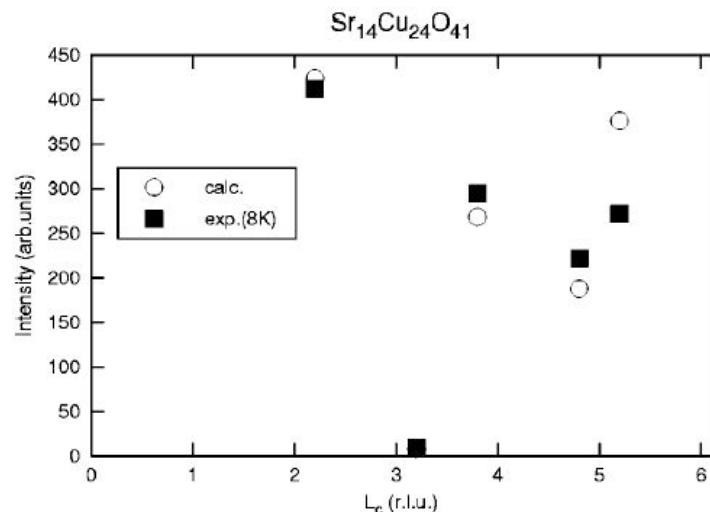
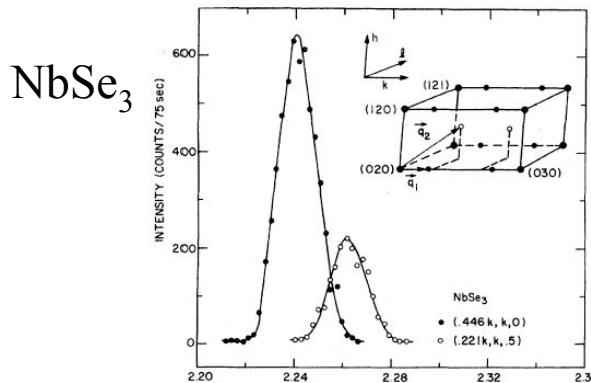


- $x = 13.6 \Rightarrow$ superconductivity $T_c = 12$ K at $P = 3$ GPa ²
- $x = 0 \Rightarrow$ insulating with a CDW ³
- $m^* \sim 50$ ($10^3 - 10^4$ more typical) ⁴
- Exhibits characteristics of *both* phases predicted by Dagotto (1992)

Study with x-ray scattering?

R. M. Fleming, D. E. Moncton, and D. B. McWhan,
Phys. Rev. B, **18**, 5560 (1978)

1. modulation wavelength (commensurate?)
2. coherence length
3. form factor (sinusoidal?)
4. $D(T)$ (mean field or no?)



T. Fukuda, *PRB*, **66**, 12104 (2002)

[refinement of study by D. E.
 Cox, *PRB*, **57**, 10750 (1998)]

l_c	L	(l, m)
1.5	15	(-2, 5)
2.2	22	(-2, 6)
3.2	32	(-1, 6)
3.8	38	(1, 4)
4.8	48	(2, 4)
5.2	52	(1, 6)

S. van Smaalen, *PRB*, **67**, 26101 (2003)

Conclusion: no obvious evidence for a CDW in Sr₁₄Cu₂₄O₄₁

Two types of CDWs

	Peierls CDW	Wigner crystal [E. Wigner, <i>Phys. Rev.</i> , 46 , 1002 (1934)]
<i>Examples</i>	NbSe_3 , $\text{K}_{0.3}\text{MoO}_3$	${}^3\text{He}$ surface, 2DEG, (Mott state!)
<i>Mechanism</i>	H_{ep}	Coulomb
<i>Effective mass</i>	$\sim 10^3 - 10^4$???
<i>Charge modulation</i>	$\sim Z = 10^1 - 10^2$	$\sim U \cdot r (E_F) \sim 10^{-2}$
<i>Cross section</i>	$\sim Z^2 \sim 10^2 - 10^4$	$\sim 10^{-4}$
<i>Bottom line</i>	Easy to measure	Hard to measure

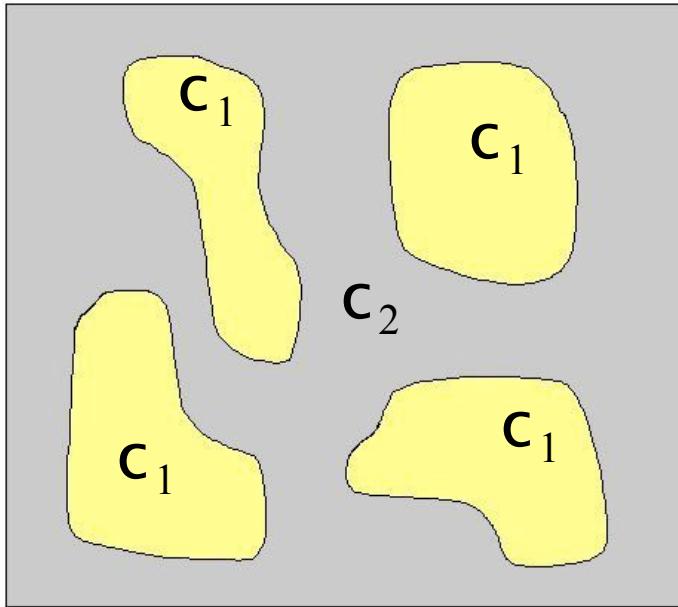
Scattering from Wigner crystal *nominally* weaker by $\sim 10^{-6}$

Hole crystal predicted by Dagotto et. al. is Wigner, not Peierls.

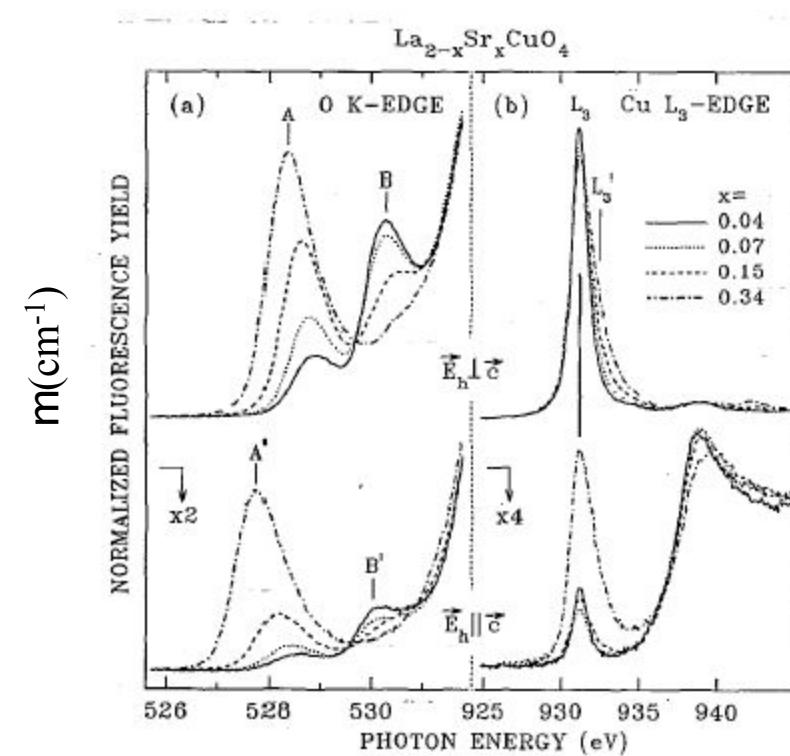
Could a Wigner crystal (many-body CDW) be responsible for the transport properties of $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$?

Resonant soft x-ray scattering

P. Abbamonte, L. Venema, A. Rusydi, G. A. Sawatzky, G. Logvenov, and I. Bozovic, *Science*, **297**, 581 (2002)



$$\mathbf{D}^{(1)} = D_0 \frac{k^2 e^{ikr}}{4\pi r} \epsilon_f \cdot \epsilon_i \int d\mathbf{x}' e^{i\mathbf{q} \cdot \mathbf{x}'} \chi(\mathbf{x}')$$

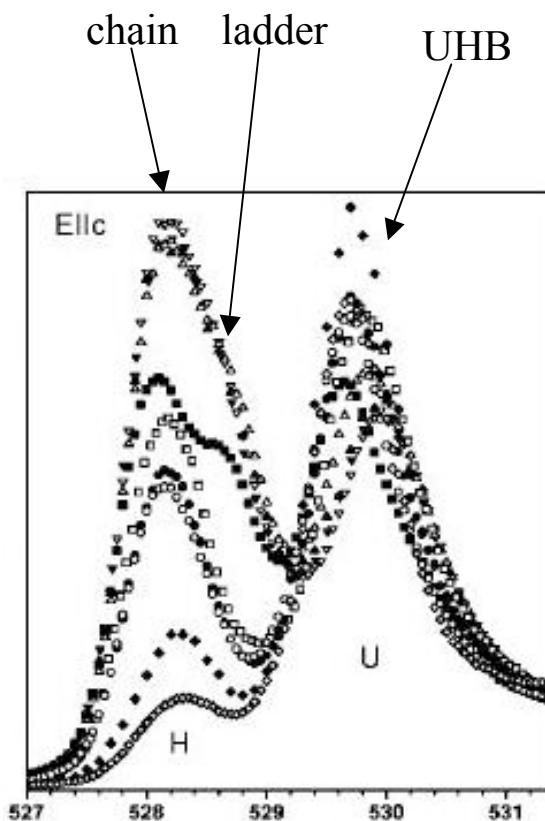
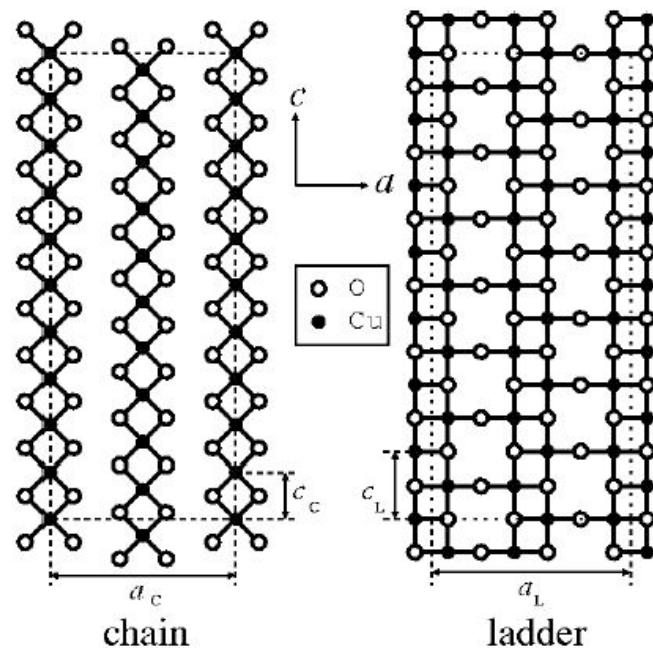


C. T. Chen, *et. al.*, PRL, **66**, 104 (1991)

$$I_{\text{normal}} \sim | \mathbf{r}(\mathbf{q}) |^2$$

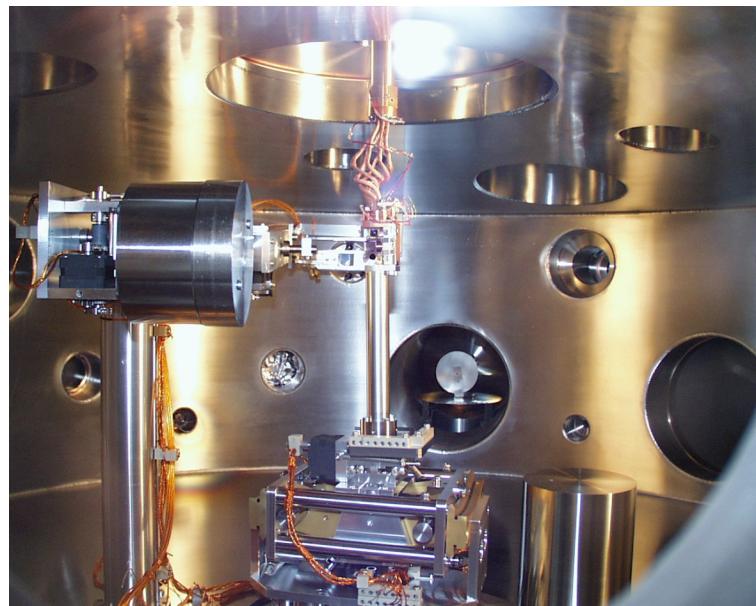
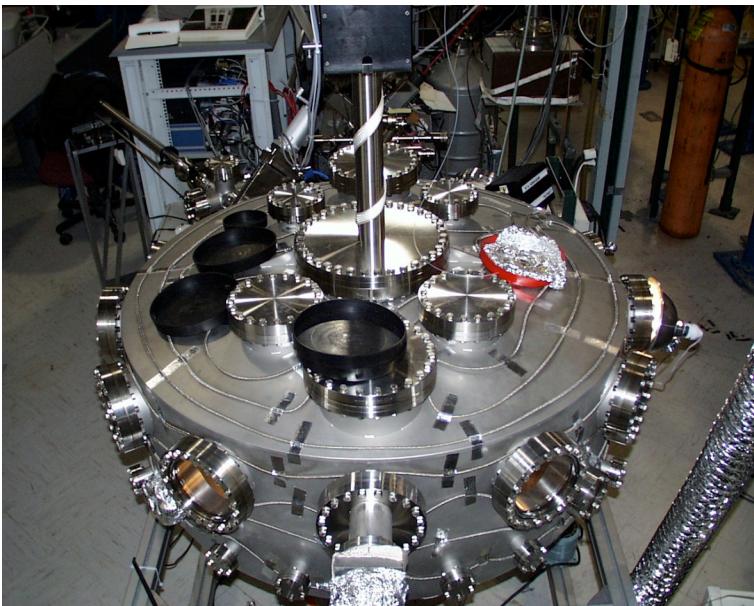
$$I_{\text{RSXS}} \sim | \mathbf{r}(\mathbf{q}) + \mathbf{a} n(\mathbf{q}) |^2 \quad n_i = S_s c_{is}^\dagger c_{is} - 1 \quad \mathbf{a} \sim 10^2$$

Edge structure in $Sr_{14}Cu_{24}O_{41}$



N. Nücker, et. al., *PRB*, **62**, 14384 (2000)

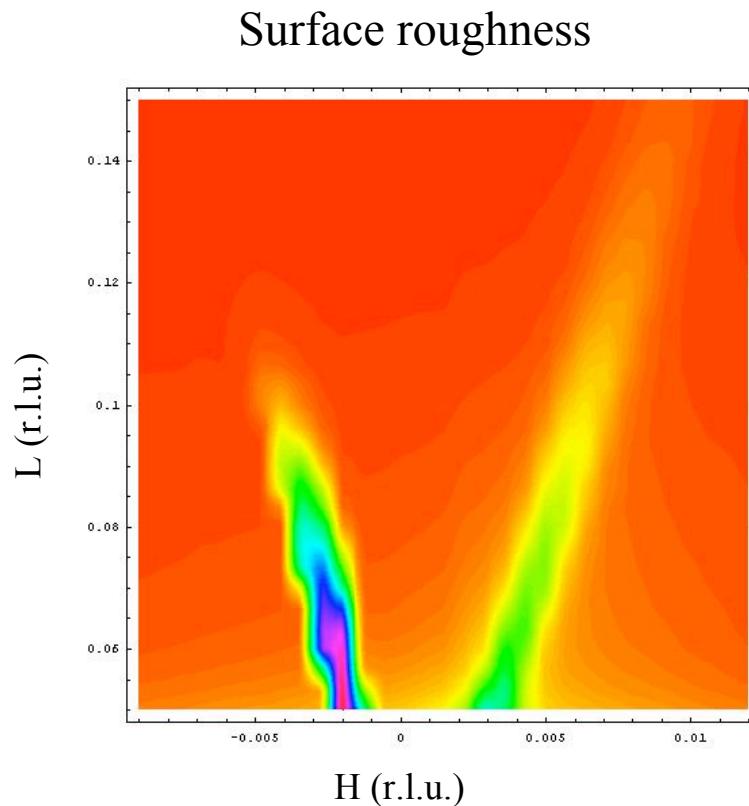
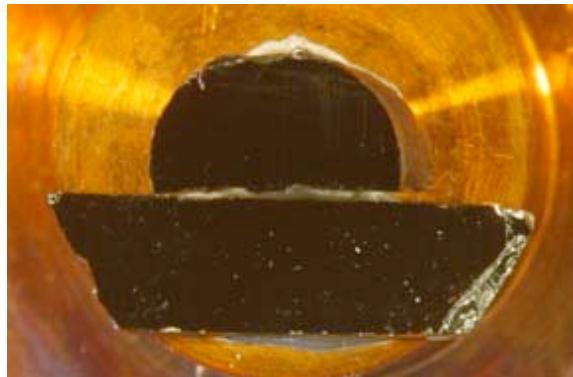
X-ray scattering in vacuum



- 1.2 m vacuum chamber
- 4 circle geometry
- Multilayer fluorescence rejection
- Channeltron / Au·CsI cathode
- He flow cryostat
- 5 Tesla magnet (vertical field)
- Base pressure = 5×10^{-10} mbar
- National Synchrotron Light Source, X1B

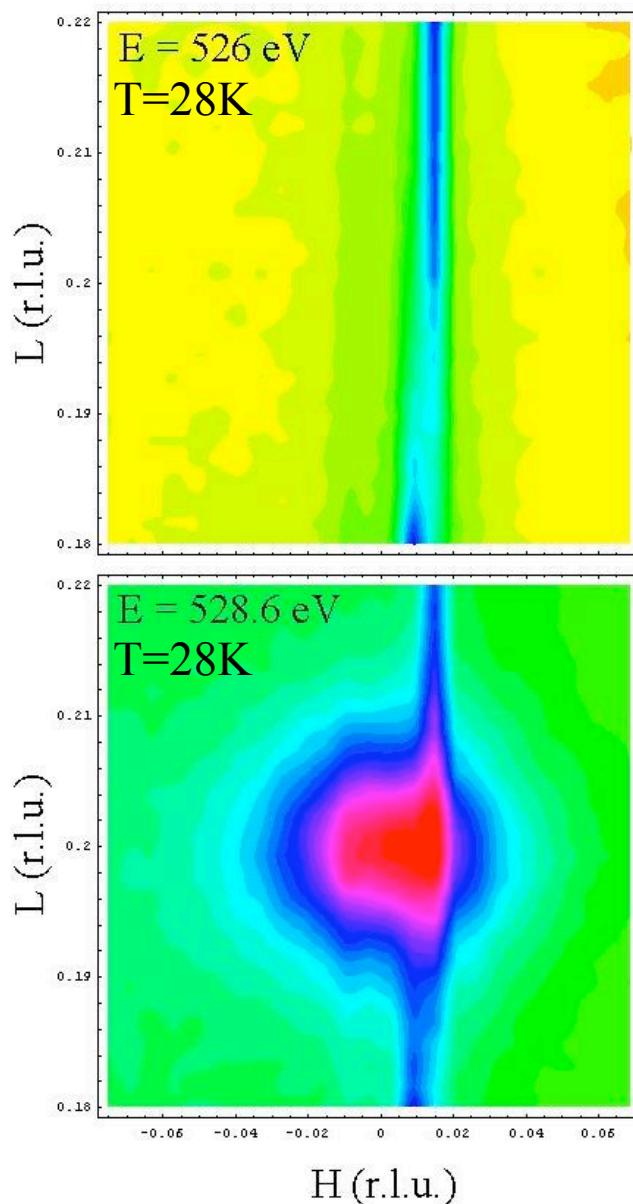
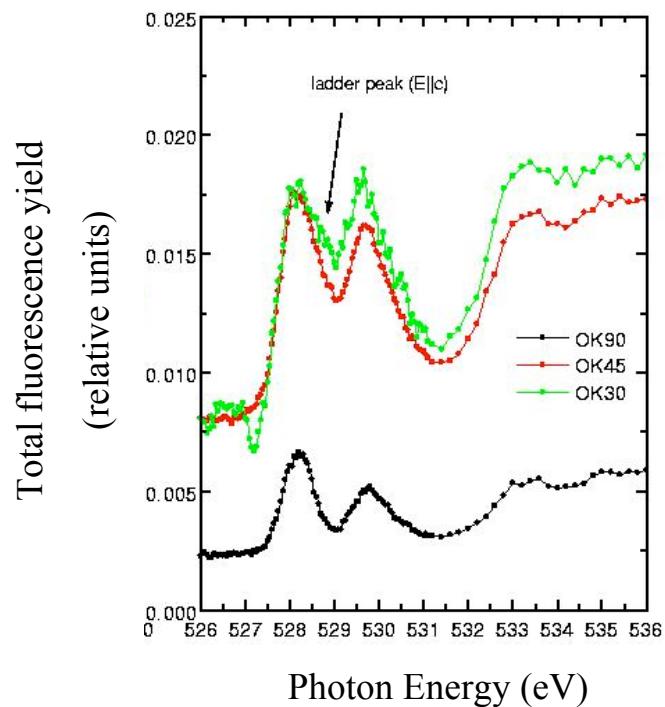
Sample prep. for low energy scattering – the surface is everything

- Traveling solvent floating zone technique*
- Polish with dry, diamond film, 30 nm \Rightarrow 10 nm $\Rightarrow \dots$ 0.1 nm
- Anneal at 120 °C in O₂ for ~1 day
- No bragg peaks! \Rightarrow Orient with Ge(111) reference sample and use surface reflection



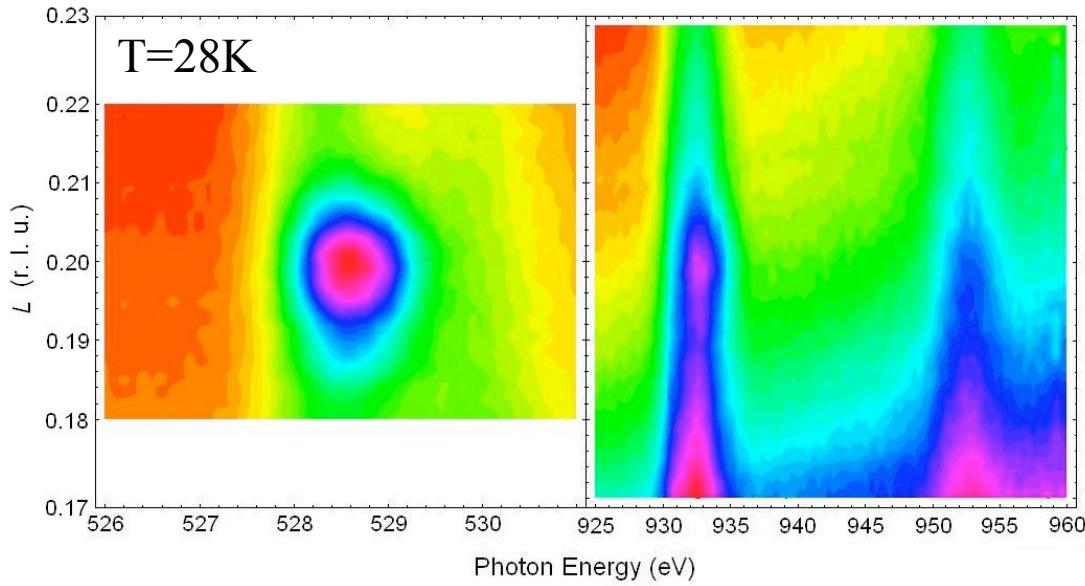
* T. Osafune, N. Motoyama, H. Eisaki, S. Uchida, *PRL*, **78**, 1980 (1997)

Valence modulation in $Sr_{14}Cu_{24}O_{41}$

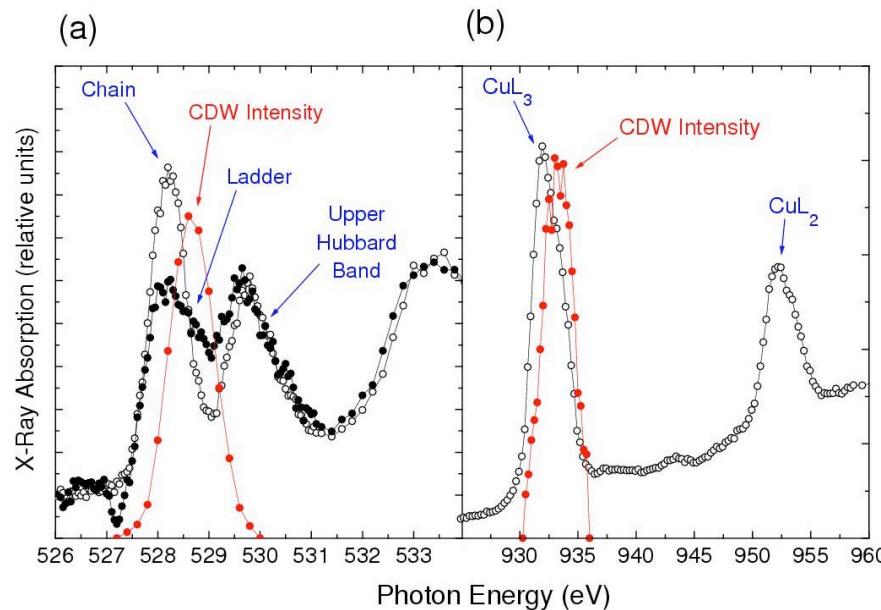


- $L = 0.200 \pm 0.009 \text{ r. l. u.} \Rightarrow I = 5.00 \pm 0.24 c_L$.
- *Does not index to 27.3 Å unit cell.*
- $x_c = 255 \text{ Å}, x_a = 274 \text{ Å}$
- No measurable off-resonant signal \Rightarrow purely electronic phenomenon

Resonance properties



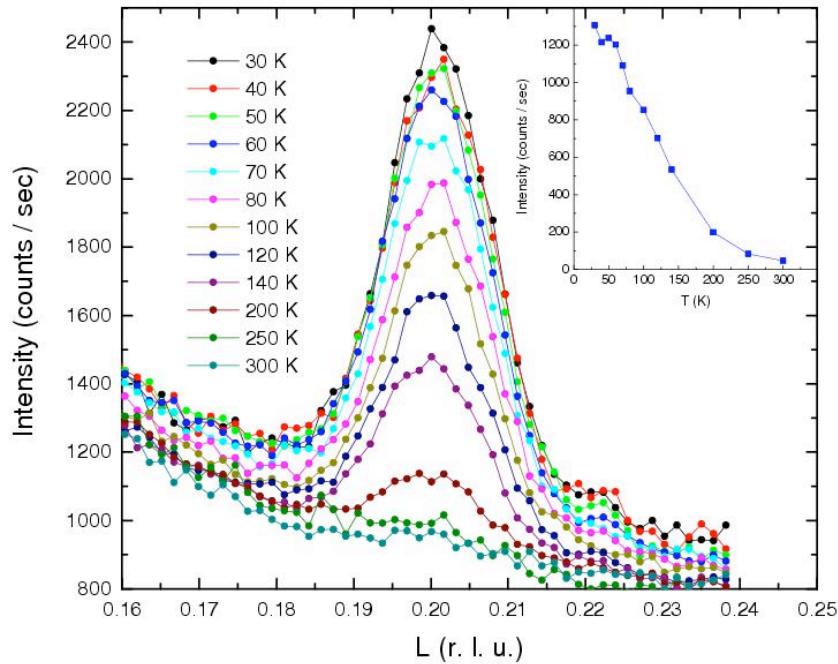
- Disappears of O_K prepeak
—*cannot be structural*
- Visible at $\text{Cu}L_{2,3}$ – still at $L=0.2$
- No harmonic at $L=0.4$ – sinusoidal



- Resonates *only* with ladder feature
- Resonates at $\text{Cu}L_3'$, not L_3 (just electrostatic)

Simplest explanation: hole crystal in the ladder, as predicted by Dagotto *et. al.*

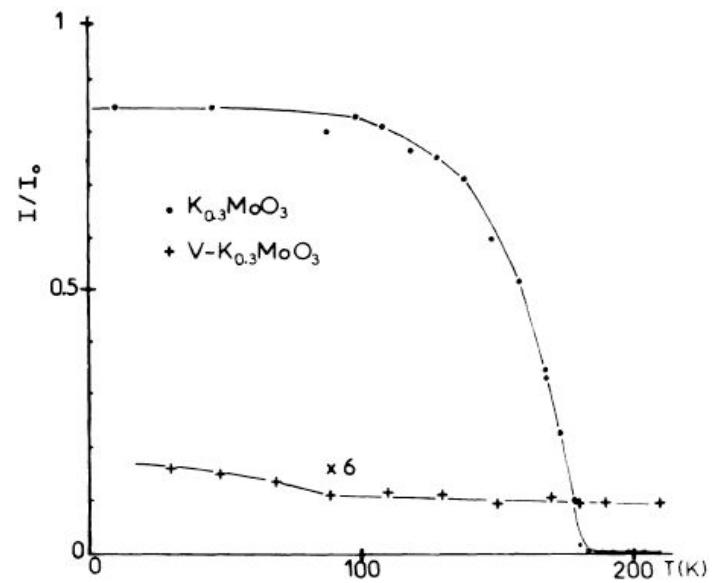
Temperature dependence



Simplest explanation: phase fluctuations
from impurities

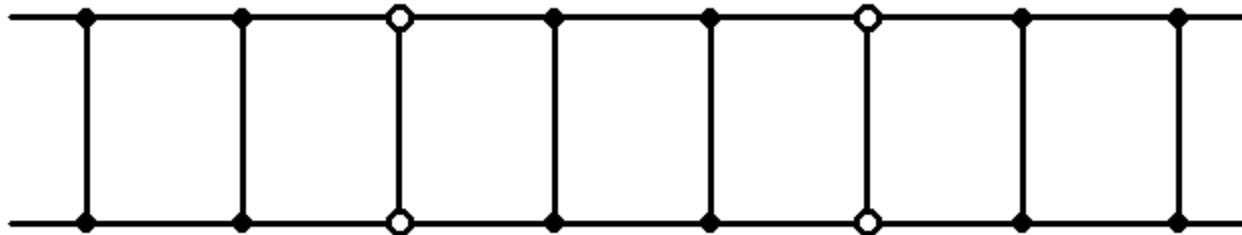


- $D(T)$ non-MFT (crossover)
- Agrees with Vučetić et. al.
[PRL, 90 257002, (2003)]
- x_c is T-independent



S. Girault, A. H. Moudden, J. P. Pouget,
J. M. Goddard, *PRB*, **38**, 7980 (1988)

Does the wavelength $\lambda = 5.00 c_L$ make sense?



- 0.8 holes in 7 rungs $\Rightarrow d = 0.8 / 14 = 0.057$
- Spin gap \Rightarrow bosonic pairs
- Expected $\lambda = 1/d = 17.54 c_L$ – *what is going on?*

Possible explanations:

- Hole density from Nücker *et. al.* incorrect
- Umklapp strong enough to draw extra charge from chains (Marston & Troyer)
- 3rd harmonic stabilized instead of 1st
- Coherent across ~ 50 neighboring ladders. Two-dimensional?

Summary

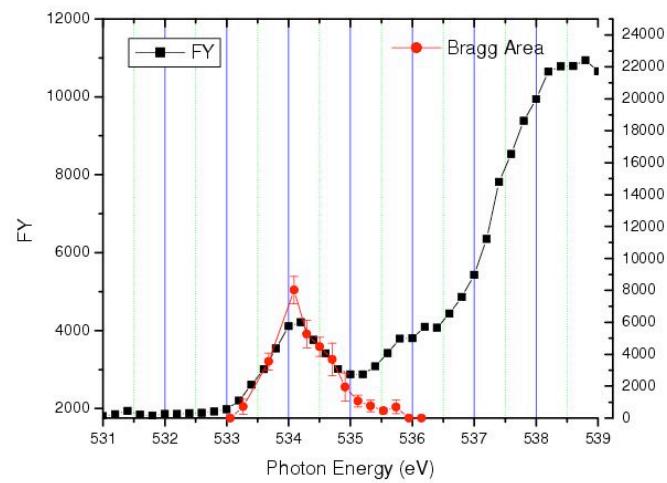
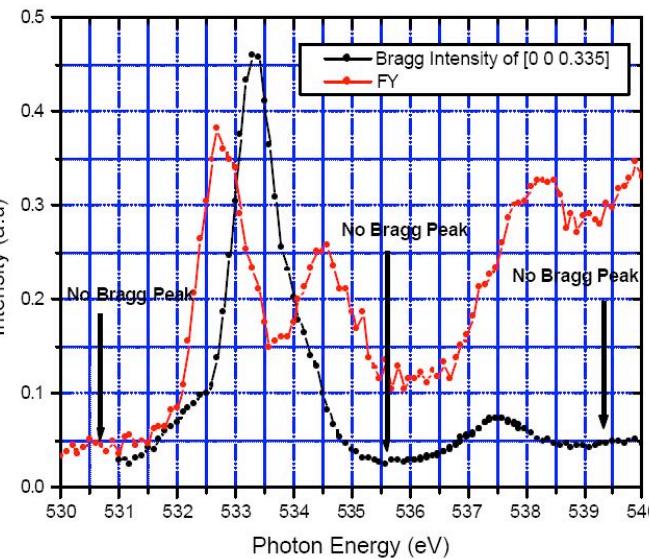
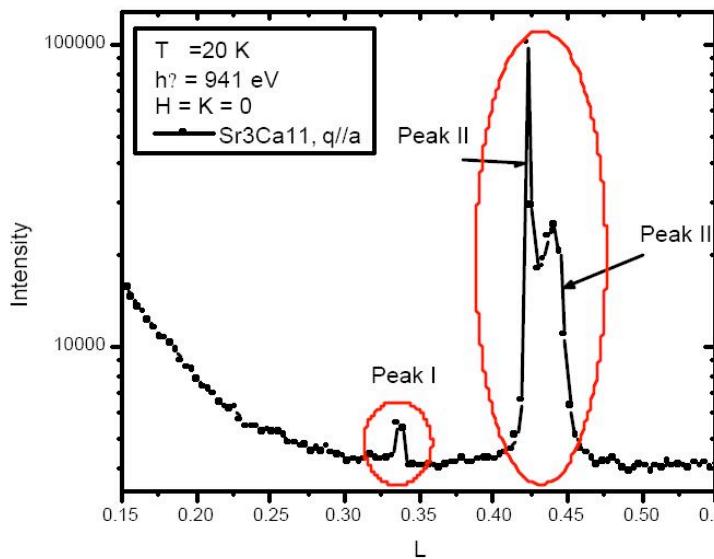
- We see a standing wave in the hole density in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$
- commensurate / resonates with ladder \Rightarrow originates in ladder
- no (measurable) lattice distortion \Rightarrow Driven by many-body interactions
- No visible harmonics \Rightarrow sinusoidal (weak-coupling *c.f.* White *et. al.*)
- $D(T)$ and $x_c(T)$ \Rightarrow impurities (sample inhomogeneity)

*Confirmed the prediction of Dagotto, Riera, Scalapino (1992)
of hole crystallization in doped spin ladders.*

Conclusions

- Explains observed CDW in transport with no Peierls distortion
- Explains low effective mass estimated by Vuletić *et. al.*
- Supports picture that superconductivity in copper-oxides occurs in close proximity (in the RG sense) to charge order

More things:



Technical notes

1. High energy
2. Emission spectrometer